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Ottawa, Canada
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INFO-0363-1

PHYSICAL CHARACTERISTICS AND SOLUBILITY OF LONG-LIVED AIRBORNE PARTICULATES IN URANIUM PRODUCING AND MANUFACTURING FACILITIES

A report upon completion of the EXTENSION to PHASE I of the study

by

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A research report prepared for the
Atomic Energy Control Board
Ottawa, Canada

Project No. 4.103.2

August 1989

Canada

Research report

91-03306/

PHYSICAL CHARACTERISTICS AND SOLUBILITY OF LONG-LIVED AIRBORNE PARTICULATES IN URANIUM PRODUCING AND MANUFACTURING FACILITIES - PHASE I EXTENSION

Experimental work carried out and report prepared by R. Robertson and S.D. Jones of Becquerel Laboratories Inc. under subcontract to MacLaren Plansearch Inc. on behalf of the Atomic Energy Control Board.

ABSTRACT

The rate of dissolution in simulated lung fluid of uranium from three different ore matrices and from refined uranium dioxide has been studied in an essentially all teflon extraction apparatus. The rate of dissolution of thorium from a nickel metal/2% thoria mixture has been similarly studied. The effect of neutron irradiation of the studied materials upon the above dissolution rates has been investigated. The adsorptions of the radioisotope tracers barium 133 (substitute for radium), lead 203 and thorium 234 upon the extraction system components has been examined.

It has been found that the total amount of uranium dissolved from -400 mesh (< 38 micron) 50-60 mg samples of CANMET BL-5, CANMET DH-1a and Mid-West uranium ores is reproducible ± 20% over six day extraction periods. For individual lung fluid fractions taken at 1 day intervals the reproducibility is much poorer. It has been found that the thoria component of the nickel/thoria material is essentially totally insoluble. No thorium was detected in any of the six lung fluid fractions collected and the total thorium removed from 50 mg samples under the experimental conditions was < 0.5 % of the thorium present in the starting material.

Neutron irradiation of the uranium ores and of uranium dioxide for two minutes under cadmium using an epicadmium neutron flux of about 5×10^{11} n cm^{-2} s^{-1} increases uranium solubility by 5% to 35% depending on the sample material. There was no observable effect on the nickel/thoria material.

The tracer work has indicated that barium 133 (and therefore presumably radium) is not adsorbed on extraction system components and is not or only moderately adsorbed or co-precipitated on the sample materials themselves. Lead 203 is partially retained within the extraction cells apparently through adsorption on the teflon walls. Only about 70% of injected lead 203 was recovered over six days from totally blank cells or cells containing only glass fibre filter material. The addition of about 50 mg of the ore DH-1a to the extraction cells changed the release pattern of lead 203 but the amount released remained at about 70% over six days. The addition of about 50 mg of the nickel/thoria material to the cells reduced the amount of lead 203 recovered to about 45% over six days. Thorium 234 showed a pattern of behaviour similar to that of lead 203, but the recoveries were lower: in the range 30%-50% and down to only 6-7% with the addition of 50 mg of nickel/thoria to the extraction cells.

RÉSUMÉ

Le présent rapport fait état des résultats de quatre analyses différentes : le débit de dissolution de l'uranium dans un fluide pulmonaire simulé à partir de trois matrices de minerai différentes et à partir de bioxyde d'uranium raffiné dans un appareil d'extraction fait essentiellement de téflon; le débit de dissolution du thorium à partir d'un mélange de nickel métal à 2 pour 100 de teneur en bioxyde de thorium; l'effet de l'irradiation neutronique des matières examinées dans des conditions de débit de dissolution indiqués ci-dessus, et les adsorptions de traceurs de baryum 133 (substitut du radium), de plomb 203 et de thorium 234 sur les composants du système d'extraction.

On a découvert que la quantité totale d'uranium dissoute à partir d'échantillons de 50 à 60 mg de minerai d'uranium de maille -400 BL-5 de CANMET BL-5, DH-1a de CANMET et de Mid-West pouvait être reproduite à 20 pour 100 plus ou moins en période d'extraction de six jours. Dans le cas des fractions de fluide pulmonaire prises à des intervalles quotidiens, la reproductibilité est beaucoup moindre. On a découvert que le bioxyde de thorium dans le mélange de nickel et de bioxyde de thorium est foncièrement tout à fait insoluble. On n'a décelé aucune trace de thorium dans aucune des six fractions de fluide pulmonaire recueillies et la quantité totale de thorium prélevée des échantillons de 50 mg dans des conditions expérimentales atteignait jusqu'à 0,5 pour 100 du thorium présent dans la matière de départ.

L'irradiation neutronique des minerais d'uranium et du bioxyde d'uranium par du cadmium durant deux minutes à l'aide d'un flux de neutron d'épicadmium d'environ 5×10^{11} n cm^{-2} s^{-1} augmente la solubilité de l'uranium de 5 à 35 pour 100 selon la nature de l'échantillon. On n'a observé aucun effet sur le mélange de nickel et de bioxyde de thorium.

L'analyse des traceurs a indiqué que le baryum 133 (et vraisemblablement le radium) n'est pas adsorbé sur les composants du système d'extraction ou seulement adsorbé modérément ou co-précipité sur les échantillons eux-mêmes. Le plomb 203 est partiellement retenu à l'intérieur des cellules d'extraction apparemment par adsorption sur les murs de téflon. Seulement 70 pour 100 environ du plomb 203 injecté ont été récupérés à partir des cellules vierges ou des cellules contenant uniquement des filtres de fibre de verre durant les six jours. L'addition d'environ 50 mg de minerai DH-1a aux cellules d'extraction a modifié le modèle de rejet du plomb 203, mais la quantité rejetée est demeurée aux alentours de 70 pour 100 durant les six jours. L'addition de quelque 50 mg de la matière faite d'un mélange de nickel et de bioxyde de thorium aux cellules a réduit la quantité de plomb 203 récupérée à environ 45 pour 100 durant la même période. Le thorium 234 a démontré un comportement semblable à celui du plomb 203, mais les récupérations ont été moins élevées, c'est-à-dire aux environs de 30 à 50 pour 100 et seulement de 6 à 7 pour 100 si l'on ajoutait 50 mg du mélange de nickel et de bioxyde de thorium aux cellules d'extraction.

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TABLE OF CONTENTS

ABSTRACT i

DISCLAIMER ii

A. INTRODUCTION 2

B. SAMPLES PROCESSED 2

C. NEUTRON IRRADIATIONS 3

D. EXTRACTION EQUIPMENT AND PROCEDURES 4

1. Teflon substitution 4
2. pH control 4
3. Temperature control 4
4. Flow rate 5

E. THE EXPERIMENTS 5

1. Experiment 1 - Unirradiated materials 5
2. Experiment 2 - Unirradiated materials 5
3. Experiment 3 - Irradiated materials 6
4. Experiment 4 - Irradiated materials 6
5. Experiment 5 - Tracer adsorption study 6

F. FIGURES AND TABLES 7

G. SUMMARY OF RESULTS AND DISCUSSION 15

H. COMMENTS AND RECOMMENDATIONS 16

1. Data Reproducibility 16
2. Neutron irradiation 16
3. Adsorption Effects 17

REFERENCES 18

A. INTRODUCTION

In PHASE I (reference 3) of this project, three areas of concern were identified by the Review Panel. The present work addresses those concerns and was therefore designed to:
a) study analytical variance, ie. the reproducibility of solubility curves derived for various materials using the developed methodology.
b) test a new procedure for neutron activation analysis of aerosol samples on filters for uranium and the effect of the neutron exposure on sample solubility.
c) test a new smaller all-teflon extraction apparatus with respect to elemental adsorption effects.

The Review Panel also recommended automatic pH monitoring and control be installed on the extraction equipment and this has been done. The panel also recommended that solubility analyses for uranium only be carried out with regard to the analytical variance study and that analyses for other elements be deferred.

This report summarises the procedures and results of a series of experiments designed to fulfil the above requirements.

B. SAMPLES PROCESSED

Five different sample materials were subjected to extraction experiments. Each was studied in unirradiated and irradiated form in triplicate to yield a total of six extraction data sets for each material.

1. Uranium ore BL-5

Canada Centre for Mineral and Energy Technology (CANMET) certified reference material BL-5 is essentially a low-grade concentrate from Beaverlodge, Saskatchewan. It has certified values for uranium 7.09%, for radium-226 857 Bq/g and for lead-210 866 Bq/g.

The BL-5 material as supplied was screened and only the -400 mesh fraction (ie. less than 38 micron) was used in the present experiments. Analysis by neutron activation of this fraction on three undried subsamples yielded uranium values of 7.23%, 7.24% and 7.49%.

2. Uranium Ore DH-1a

CANMET certified reference material DH-1a is a uranium ore typical of the Elliot Lake area in Ontario. It has certified values for uranium 0.2629%, thorium 0.091%, radium-226 31.5 Bq/g and lead-210 30.8 Bq/g. Separate analyses by Becquerel Laboratories Inc. for thorium-234 and thorium-230 yielded values of 30.8 and 28.7 Bq/g respectively indicating secular equilibrium for this material. The physical form of DH-1a is a powder of -200 mesh, ie. less than 75

micron.

The DH-1a material as supplied was screened and only the -400 mesh fraction was used in the present experiments. Analyses by neutron activation of this fraction on three undried subsamples yielded uranium values of 0.215%, 0.194% and 0.212%.

3. Uranium ore from the Canadian mid-west (MWO)

MWO is a medium grade ore from Saskatchewan. It was the material studied in the feasibility study carried out prior to this work (see reference 1). A screened -400 mesh particle size fraction was taken. Analysis by neutron activation of this fraction on three undried subsamples yield uranium values of 2.57%, 2.62% and 2.51%.

4. Uranium dioxide

The fourth material studied was unsintered uranium dioxide commercially produced by the fluid bed process. The physical form of this material was a very dark brown, almost black powder of which only a -400 mesh fraction was used. Analysis by neutron activation of four dried subsamples of this fraction yielded values for uranium of 86.3%, 84.7%, 86.6% and 87.3%, which on the average is only about 92% of the theoretical uranium value based on UO_2 stoichiometry.

5. Thoria/nickel

The fifth material studied was a fine powder mixture of thorium dioxide and nickel metal. The source of this material was an industrial process in western Canada. The nominal constituent concentrations were thorium dioxide 2% and nickel 98%. No elemental analysis of the material was carried out by Becquerel Laboratories Inc. It was used as supplied: a black powder of unspecified particle size distribution.

C. NEUTRON IRRADIATIONS

For that part of the study involving extraction experiments on neutron irradiated samples, 0.5g specimens of each of the five materials described in section C above were irradiated in the 2MW pool reactor at McMaster University. The irradiations were carried out in an epithermal neutron position at a cadmium shielded terminus of a pneumatic insertion facility (rabbit). The epicadmium flux was about 5×10^{11} n cm^{-2} s^{-1}. The duration of the irradiations was 2 minutes.

The conditions of irradiation were chosen as an estimate of the minimum exposure required to effectively determine uranium on filters by neutron activation analysis at the 10 microgram level with a detection limit of 100 nanograms.

D. EXTRACTION EQUIPMENT AND PROCEDURES

The extraction equipment was changed in several ways relative to the system previously used (See Figure 1).

1. Teflon substitution

All parts of the extraction equipment including and after the extraction cell were converted to teflon as far as possible. The single exception was the membrane filter. This was a Millipore Corporation product made of their Durapore (polyvinylidene diflouride) material. The filter description was HVLP04700 Durapore hydrophilic 47mm 0.45 micron. The upstream side including the simulated lung fluid reservoir and the lines leading to the extraction cells and connections was of other plastic materials.

The extraction cells were 15mL screw cap teflon bottles. These were connected by ¼ inch teflon tubing on the outlet side to all-teflon filter holders. These two-part filter housings were commercially supplied and initially were unsatisfactory because of leakage of fluid around the edge of the filter. They were modified through a design change which eliminated the screw type closure and substituted a direct press fit of the two parts through application of a powerful clamp. This modification avoided the creasing of the filter at the edges caused by the former twisting action of the closure and allowed much greater pressure to be applied.

The outlet sides of the filter holders were connected by ¼ inch teflon tubing through all-teflon needle valves to polypropylene collection bottles.

2. pH control

In previous work connected with this project (references 1,2,3), the pH of the input simulated lung fluid was controlled manually through sampling and adjustment. In the present work, this function was carried out automatically using a Sentry 2000 control system supplied by XYMOTEC BIOSYSTEMS Inc. of Montreal. This unit consisted of a pH electrode sensor in the lung fluid reservoir with feedback to an electronics module which included a peristaltic pump which was activated as required to deliver 1N HCl to the lung fluid reservoir. After some electronic modification, this unit worked quite reliably and controlled the input lung fluid pH to 7.30±0.05.

3. Temperature control

While running, all of the extraction system components were maintained at 37°C within a single plywood box with a clear acrylic plastic window. The box comfortably accommodated 8 systems working in parallel. Heating was supplied through a thermostatically controlled resistance type baseboard heater. The air inside the box was constantly and vigorously circulated by two fans. The

temperature was monitored through visual checks using an ordinary thermometer and found to be very steady at 37±1°C.

The above constant temperature enclosure replaced the previous box within a box concept.

4. Flow rate

For all of the experiments in this study, a (approximately) constant flow regime was adopted. Fractions of about 225mL were collected at 24 hour intervals over a six day period. At the end of the collection period, fractions were acidified with 2mL concentrated hydrochloric acid prior to storage.

In previous work, the flow rate was allowed to decrease with time.

E. THE EXPERIMENTS

Five experiments each involving eight parallel extractions were run. The first four involved normal extractions to study the solubility of the five materials (irradiated and unirradiated) described in Section B. The fifth experiment was a tracer experiment designed to study adsorption effects. In all cases, the experiments were run over six day periods and six 24h fractions of lung fluid were collected. Lung fluid fraction volumes were typically in the 200-250mL range.

1. Experiment 1. (Unirradiated materials)

Cell	1.	BL-5	57mg	(sample plus 25mm glass fibre filter)
	2.	BL-5	51mg	"
	3.	DH-1a	57mg	"
	4.	DH-1a	55mg	"
	5.	MWO	57mg	"
	6.	MWO	57mg	"
	7.	U-dioxide	58mg	"
	8.	Blank	-	(25mm glass fibre filter only)

2. Experiment 2. (Unirradiated materials)

Cell	1.	U-dioxide	58mg	(sample plus 25mm glass fibre filter)
	2.	U-dioxide	53mg	"
	3.	Ni/Thoria	58mg	"
	4.	Ni/Thoria	51mg	"
	5.	BL-5	56mg	"
	6.	DH-1a	50mg	"
	7.	MWO	52mg	"
	8.	Ni/Thoria	59mg	"

3. Experiment 3. (Irradiated materials)

Cell	1.	BL-5	56mg	(sample plus 25mm glass fibre filter)
	2.	BL-5	57mg	"
	3.	DH-1a	50mg	"
	4.	DH-1a	57mg	"
	5.	MWD	57mg	"
	6.	MWD	52mg	"
	7.	U-dioxide	59mg	"
	8.	Blank	-	(25mm glass fibre filter only)

4. Experiment 4. (Irradiated materials)

Cell	1.	U-dioxide	53mg	(sample plus 25mm glass fibre filter)
	2.	U-dioxide	52mg	"
	3.	Ni/Thoria	53mg	"
	4.	Ni/Thoria	61mg	"
	5.	BL-5	55mg	"
	6.	DH-1a	57mg	"
	7.	MWD	54mg	"
	8.	Ni/Thoria	58mg	"

5. Experiment 5. (Tracer adsorption study)

Cell	1.	Complete blank	-	(no sample, no filter)
	2.	Complete blank	-	"
	3.	Filter blank	-	(25mm glass fibre filter only)
	4.	Filter blank	-	"
	5.	DH-1a	52mg	(sample only)
	6.	DH-1a	51mg	"
	7.	Ni/Thoria	56mg	"
	8.	Ni/Thoria	54mg	"

Experiment 5 was run as a normal experiment except that 1mL of a mixed tracer solution was added to the extraction cells and allowed to equilibrate with the cell contents for 30 minutes prior to flow commencement. The tracer solution was a mixture of the carrier-free isotopes barium-133 (substitute for radium-224), lead-203 and thorium-234 at pH 3. The solution was filtered through a 0.22 micron filter immediately before use. Prior tests indicated that the addition of 1mL of this solution to 15mL (extraction cell volume) of simulated lung fluid resulted in a pH decrease from 7.3 to 7.2. The lung fluid fractions collected were all made up to 250mL for direct comparison by gamma counting of 1mL of the tracer solution similarly diluted.

F. FIGURES AND TABLES

Figure 1 is found on page 7. Tables 1-7 are found on pages 8-13.

F. FIGURES AND TABLES

Figure 1: Schematic Representation of Extraction Equipment

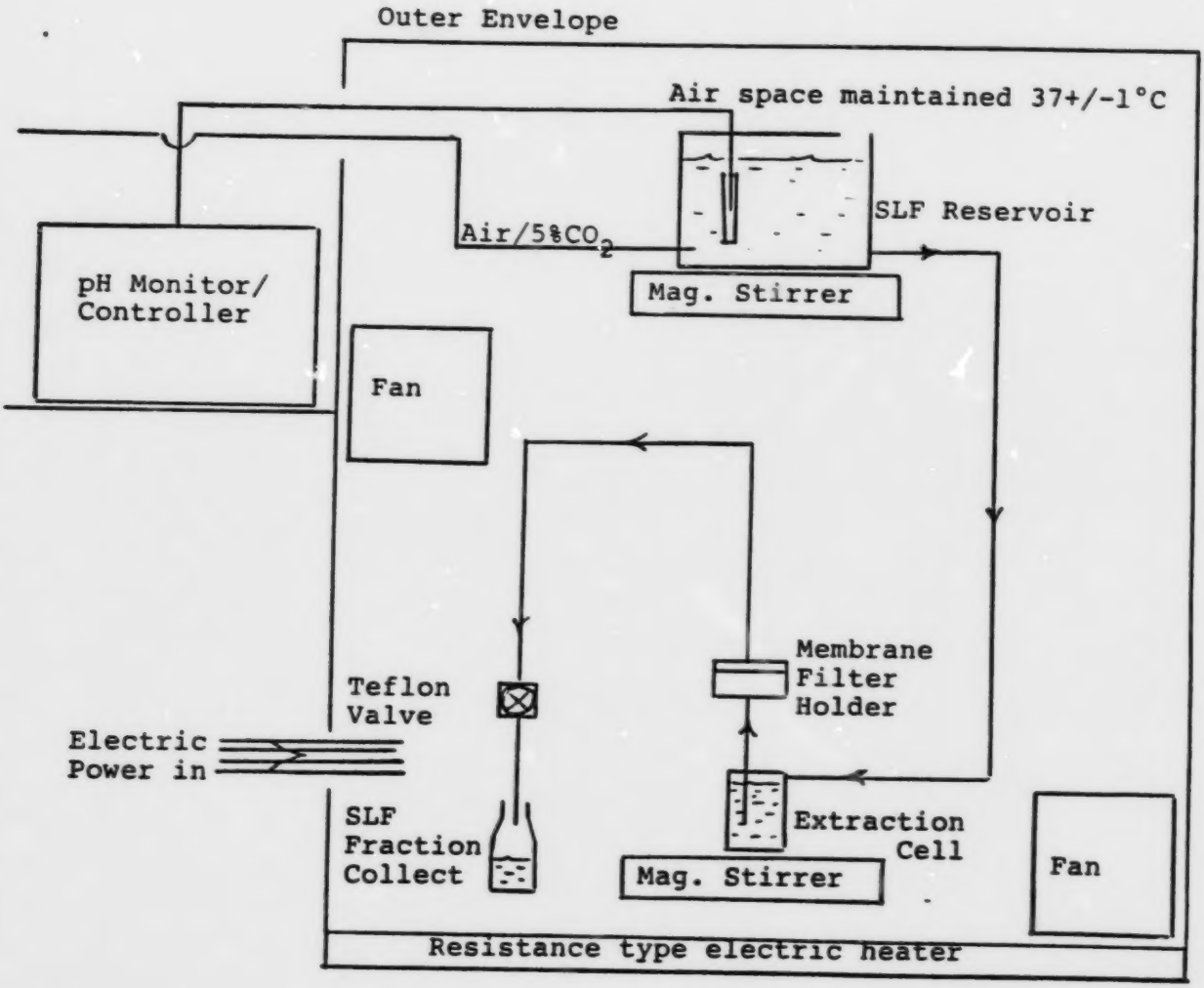

Table 1: Extraction of uranium from BL-5 uranium ore

	Fraction of Available Uranium Extracted (%)					
Lung Fluid Fraction	Unirradiated material			Irradiated material		
	Expt. 1	Expt. 1	Expt. 2	Expt. 3	Expt. 3	Expt. 4
1.	4.6	3.3	5.1	5.0	4.1	5.1
2.	5.8	4.8	3.4	7.3	7.8	5.6
3.	4.5	3.3	3.2	4.5	3.7	4.2
4.	3.5	2.7	3.3	3.5	3.1	4.1
5.	2.7	3.2	2.4	3.2	2.8	4.0
6.	2.5	2.5	2.9	2.9	2.6	3.8
Total U extracted	23.6%	19.8%	20.3%	26.4%	24.1%	26.8%
Mean of 3 extractions		21.2%			25.8%	

Note: BL-5 -400 mesh fraction taken to be 7.32% U.

Table 2: Extraction of uranium from DH-1a uranium ore

	Fraction of Available Uranium Extracted (%)					
Lung Fluid Fraction	Unirradiated material			Irradiated material		
	Expt. 1	Expt. 1	Expt. 2	Expt. 3	Expt. 3	Expt. 4
1.	17.5	10.5	18.1	21.0	20.8	18.6
2.	2.8	2.7	2.0	3.2	2.8	3.2
3.	1.6	1.7	1.5	0.9	1.6	1.8
4.	1.0	1.0	0.6	0.5	0.6	1.1
5.	0.4	0.4	0.8	0.5	0.6	0.8
6.	< 0.4	< 0.4	< 0.4	< 0.5	0.6	< 0.5
Total U extracted	23.3%	16.3%	23.0%	26.1%	27.0%	25.5%
Mean of 3 extractions		20.9%			26.2%	

Note: DH-1a -400 mesh fraction taken to be 0.207% U.

Table 3: Extraction of uranium from mid-western uranium ore (MWO)

Fraction of Available Uranium Extracted (%)

Lung Fluid Fraction	Unirradiated material			Irradiated material		
	Expt. 1	Expt. 1	Expt. 2	Expt. 3	Expt. 3	Expt. 4
1.	31.7	27.4	31.0	26.8	28.3	16.2
2.	19.3	19.4	15.0	23.4	22.1	23.6
3.	11.2	9.7	8.9	9.4	9.0	14.5
4.	3.8	4.4	6.0	5.0	5.2	8.1
5.	2.5	3.7	2.6	2.6	3.6	6.2
6.	1.3	1.8	3.9	1.9	2.6	3.4
Total U extracted	69.8%	66.4%	67.4%	69.1%	70.8%	72.0%
Mean of 3 extractions		67.9%			70.6%	

Note: MWO -400 mesh fraction taken to be 2.57% U.

Table 4: Extraction of uranium from uranium dioxide

Fraction of Available Uranium Extracted (%)

Lung Fluid Fraction	Unirradiated material			Irradiated material		
	Expt. 1	Expt. 2	Expt. 2	Expt. 3	Expt. 4	Expt. 4
1.	0.508	0.540	0.510	0.623	0.689	0.698
2.	0.0173	0.0263	0.0298	0.0255	0.0441	0.0386
3.	0.0095	0.0125	0.0201	0.0102	0.0256	0.0325
4.	0.0080	0.0091	0.0115	0.0074	0.0223	0.0214
5.	0.0056	0.0051	0.0065	0.0043	0.0120	0.0148
6.	0.0037	0.0065	0.0075	0.0053	0.0161	0.0089
Total U extracted	0.55%	0.60%	0.58%	0.68%	0.81%	0.81%
Mean of 3 extractions		0.58%			0.77%	

Note: Uranium dioxide -400 mesh fraction taken to be 86.2% U.

Table 5: Experiment 5 - Recovery of barium-133 tracer

	Fraction of Ba-133 recovered (%)							
	Cell 1	Cell 2	Cell 3	Cell 4	Cell 5	Cell 6	Cell 7	Cell 8
Fraction 1	105	101	95.7	100	70.4	71.9	104	101
2	0.7	0.7	0.5	0.6	5.8	5.9	< 0.3	< 0.3
3	0.3	< 0.2	0.3	0.4	2.2	2.4	< 0.3	0.3
4	< 0.3	< 0.3	0.1	0.3	1.3	1.6	< 0.3	< 0.3
5	< 0.2	0.3	< 0.3	< 0.2	1.1	1.5	< 0.2	< 0.3
6	0.2	< 0.2	0.2	< 0.3	1.2	1.0	0.3	< 0.3
	~100%	~100%	~100%	~100%	82%	84%	~100%	~100%
Cell Filter	< 0.1%	< 0.1%	< 0.1%	< 0.1%	2.1%	1.3%	< 0.1%	< 0.1%

Table 6: Experiment 5 - Recovery of lead-203 tracer

	Fraction of Pb-203 recovered (%)							
	Cell 1	Cell 2	Cell 3	Cell 4	Cell 5	Cell 6	Cell 7	Cell 8
Fraction 1	66.7	60.3	63.0	59.3	50.3	50.1	30.0	31.7
2	2.8	3.2	4.1	3.3	7.6	5.9	7.6	5.8
3	0.8	0.8	1.4	1.9	6.1	4.2	3.8	3.2
4	0.5	0.2	1.3	0.7	3.2	2.7	2.1	1.3
5	0.3	0.4	1.0	0.8	2.6	1.8	1.9	1.1
6	0.2	0.3	0.4	0.3	1.8	2.2	0.9	0.5
	71.3%	65.2%	71.2%	66.6%	71.6%	66.9%	46.3%	43.6%
Cell Filter	3.1%	4.0%	5.5%	4.4%	6.1%	4.8%	7.9%	14%

Table 7: Experiment 5 - Recovery of thorium-234 tracer

	Fraction of Th-234 recovered (%)							
	Cell 1	Cell 2	Cell 3	Cell 4	Cell 5	Cell 6	Cell 7	Cell 8
Fraction 1	30.4	36.8	33.9	33.5	48.2	37.3	3.0	4.9
2	3.0	5.3	4.3	3.7	2.2	< 1.6	1.1	< 0.5
3	2.1	1.4	2.0	1.1	< 1.1	< 1.1	1.1	< 1.1
4	< 0.8	< 0.8	1.7	1.2	< 0.8	1.1	< 0.7	1.1
5	< 0.8	1.2	1.1	1.2	1.3	< 1.2	< 0.5	< 0.6
6	< 0.8	< 0.5	1.7	1.3	1.2	1.1	< 0.8	1.3
	~36%	~45%	44.7%	42.0%	~53%	~40%	~6%	~7%
Cell Filter	20%	18%	17%	18%	20%	12%	5.3%	5.5%

G. SUMMARY OF RESULTS AND DISCUSSION

No mathematical analysis of data is attempted in this report. Such rigorous methods require more data than this work makes available. What follows below is a collection of reasonably conservative assertions based on the information that has been produced.

Tables 1-4 indicate that the total amount of uranium dissolved is reproducible within ±20% for a given material under the experimental and analytical conditions used. The data in these tables indicate, however, that corresponding individual lung fluid fractions may differ much more than this and that difference factors between 2 and 3 are not unusual in situations were the amount of uranium in a lung fluid fraction is small relative to the uranium available in the sample.

There are no data presented in tabular form for the fifth material studied. The nickel/thoria material is essentially completely insoluble in simulated lung fluid in both unirradiated and irradiated form. A total of six subsamples of 50-60mg each were subjected to six-day extractions, as for the other materials studied. Analyses of the lung fluid fractions failed to detect any thorium at a detection limit of 1 microgram per fraction. Since the concentration of thorium in the starting material is 2% it can be stated that each lung fluid fraction contained less than 0.1% of the available thorium. It is reasonable to assert that a six day extraction dissolves less than 0.5% of the available thoria.

Tables 1-4 also indicate that neutron exposure of the studied materials increases solubility, which is logical from neutron damage arguments. The observed effect, under the irradiation conditions used, ranges from about 5% to about 35% depending on the material. Table 4 indicates that post-irradiation beta/gamma damage may also be a factor, based on the different solubility observed for UO_2 in Experiment 3 relative to Experiment 4. These experiments were performed on the same irradiated material but the interval between them was about two weeks.

Tables 5-7 were constructed from the results of Experiment 5 and show the recovery of tracers injected into the extraction cells.

Barium-133 was only retarded in Cells 5 and 6 which each contained about 50mg of DH-1a uranium ore and no other material. It appears that the ore material alone is responsible for the hold-up presumably through a coprecipitation/adsorption mechanism. The release of Ba-133 from the ore appears to level out at about 1% per day by the sixth day.

Lead-203 was delayed in the extraction cells under all conditions tested. In Cells 1-6 about 70% of the tracer was recovered in lung

fluid fractions over 6 days and it would initially appear that the missing 30% is mostly adsorbed on the teflon walls of the extraction system. However, the shapes of the recovery curves for Cells 5 and 6, which contained DH-1a uranium ore, are different from the curves for Cells 1-4. It may be that elements leached from the ore displace Pb-203 from wall adsorption sites but that the ore itself offers replacement adsorption sites. In cells 7 and 8 it appears that the nickel/thoria material definitely plays a role in adsorption.

Thorium-234 was delayed in the extraction cells under all conditions tested. The observed effect was greater than that for lead-203. The data for Cells 1-4 indicate strong adsorption on the teflon cell walls. The data for Cells 5 and 6 indicate that adsorption on uranium ore material plays a role. The data for Cells 7 and 8 indicate very strong adsorption of the thorium-234 tracer on the nickel/thoria material and very slow or negligible release.

H. COMMENTS AND RECOMMENDATIONS

The data arising from this study largely speak for themselves. In the main it is the responsibility of others such as the lead contractor, the AECB steering committee and the external review panel to draw inferences from it and assess its impact upon future work on the project. The authors of this report, however, do offer the following broad comments and recommendations:

1. Data reproducibility

It seems that it will be possible, with the methodology developed to its present state, to reproduce solubility data for uranium over long term extractions to the required ±20%. The same is probably true for radium. Short term extractions of a few days or less will not be so reproducible. Very tight pH control seems to be critical.

2. Neutron irradiation

Even the much reduced neutron exposure in this study relative to previous work has a significant effect on solubility of uranium and possibly other elements. However, the effect is not so great as to preclude neutron activation as the method of choice for determining uranium and possibly thorium on the filter substrates which are to be the main subject of future work. In any case, there seems to exist no good alternative. It may well be that a good proportion of the radiation damage sustained by the samples was self-induced by post-irradiation beta/gamma radiation. Bulk samples of about 0.5g were irradiated and stored for 1-3 weeks before processing. Real samples on filters will be much smaller. Solubility processing of these should follow immediately after neutron activation analysis.

3. Adsorption effects

It appears that uranium and radium are not adsorbed on the extraction system surfaces (including the samples themselves) to an extent which would cause difficulties in interpreting long term solubility data derived through the current methodology.

The same cannot be said for thorium and lead. Thorium is strongly adsorbed and lead is significantly adsorbed and the adsorbed species are so slowly released that the interpretation of solubility data for these elements may be compromised. If there is no equivalent mechanism for lead and thorium in the real lung situation, it can be said that the solubility data derived with the current system will not be very useful.

It is recommended that lung clearance studies of thorium-234 and lead-203 tracers be carried out. Failing this, the elements lead and thorium should be dropped from this project.

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